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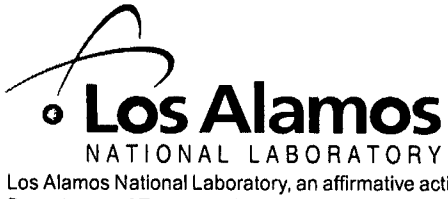

Form 836 (8/00)

Results of a Comparison Study Using Tomographic and Segmented Gamma Scanner Technology

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Abstract

In order to support the many on-going research and programmatic activities at Los Alamos' Plutonium and Chemistry and Metallurgy Research (CMR) Facilities in as accurate, efficient, and cost-effective manner possible, every reasonable effort is made to equip the nondestructive assay (NDA) laboratories with the most modern and technologically advanced instrumentation available. Recently, new state-of-the-art tomographic gamma scanner (TGS) instruments were installed to replace aging and outmoded segmented gamma scanner (SGS) instruments. Through the implementation of a translation axis, in addition to the vertical and rotation axes of the SGS, the TGS technique is able to employ axial tomography to determine the spatial distribution and quantity of nuclear material using high-resolution gamma-ray spectroscopy. Because the attenuation matrix and source distributions are known more accurately than with the SGS technology, biases due to matrix and source distributions should be reduced. In principle, a single calibration should suffice for the determination of isotopic mass for a wide range of material and matrix types. A number of questions naturally arise concerning these purported advantages of the TGS. Perhaps the most fundamental of these is to understand how the TGS measurement results compare with those of a typical SGS on the same well-characterized standards differing in matrix and material type. To that end, the TGS operating parameters were optimized to assay 55-gallon drum waste identical to that measured by our SGS. The calibration and measurement results on these standards, placed in typical low-density waste matrices, are presented and discussed. These results should enable more confident use of the TGS as well as point the way toward even more studies to enable more effective employment of the new TGS technology.

Introduction

For the past twenty-five or so years, SGS instruments[1] have been utilized at Los Alamos' Plutonium and CMR Facilities to assay uranium and plutonium waste embedded in varied low-density matrices and geometries. While the SGS technique represented a substantial improvement over the older far-field[1] approach by enabling gamma-ray attenuation corrections on a segment-by-segment basis, it never the less left much to be desired in certain measurement situations. For example, whenever inhomogeneities or voids exist within a segment, the attenuation correction will be inaccurate because the correction necessarily spans the entire segment whereas the discontinuity behaves essentially as a mathematical singularity within the segment.[2] In addition, so-called end effects are

always a problem for items differing in height from the calibration standards.[3] In principal, the TGS technology[4] corrects, or at least substantially reduces, these biases by breaking the individual segments into a series of volume elements (voxels) and attenuation-correcting each voxel. The number of voxels per segment (resolution) can be varied with optimization depending on such factors as the complexity and geometry of the item to be assayed as well as the availability of different-sized collimators and calibration standards. The end effects are greatly reduced or eliminated through utilization of layer coupling.[5]

We purchased two drum-sized TGS instruments from the ANTECH Corporation[6] to replace old and outmoded SGS instruments in our plutonium and CMR facilities at Los Alamos. The TGS located in the nondestructive assay (NDA) laboratory at CMR is shown in Figure 1. Perhaps the most striking difference between the TGS and SGS, as

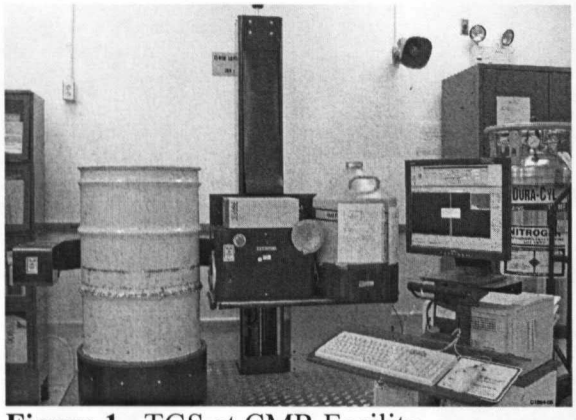

Figure 1. TGS at CMR Facility

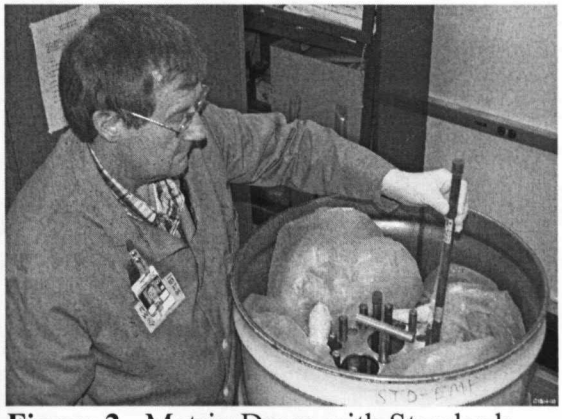

Figure 2. Matrix Drum with Standards

seen in Figure 1, is that the detector and transmission source (^{75}Se) assembly move together vertically with the platform rotating and moving horizontally. With our SGS, because the horizontal degree of freedom is not needed, the platform rotates and moves vertically with the detector and source assembly remaining fixed and motionless. The two computers, one for data acquisition and the other for analysis, can be seen in the foreground just behind and under the keyboard and monitor screen. The large 160-liter liquid nitrogen dewar seen in the background is used to fill the much smaller detector dewar. The drum in Figure 1 seen resting on the platform contains the matrix material in which the ^{238}Pu and ^{239}Pu standards are placed for calibration and measurement control assays (see Figure 2). The ^{235}U standards[7] are housed in their own slightly oversized (several inches greater in height) drums.

Preparation and Data Acquisition

We now describe the procedure for acquiring the comparison data. First, however, we had to calibrate the TGS. We have three sets (^{239}Pu, ^{238}Pu, and ^{235}U, traceable to NIST standard reference materials) of calibration standards at CMR that were made specifically[7,8,9] for the SGS and should therefore also be ideal for the TGS. The ^{239}Pu (93.78%) set consists of five "sheet" standards that span a ^{239}Pu mass range of 0.1680 grams to 4.1700 grams. The standards were constructed to distribute the PuO_2 over an area simulating PuO_2 distribution on glovebox gloves, plastic bags, and cellulose or

fabric towels typically used in plutonium-handling glovebox operations. They were also designed to distribute the PuO_2 in a layer sufficiently thin to reduce any self-attenuation effects (and corresponding transmission measurement corrections relating to the PuO_2) to a negligible value as compared to other uncertainties associated with SGS measurements.[8] The sheet standards were placed inside plastic bags that were subsequently placed inside thin-walled stainless steel overpack containers about 9 inches in diameter and 11 inches height.

The ^{238}Pu (enriched to 82.52%) standards[9] consist of 12 tubes containing 51.2 mg ^{238}Pu per tube. The ^{238}Pu was disbursed in a silica (diatomaceous earth) matrix sealed in glass tubes about 50 cm long. Each glass tube was overpacked and sealed in a 14 mm inner diameter, 58 cm long thin-walled stainless-steel tube. Twelve additional blank tubes were made and the total of twenty four were placed in a circular carousel of about 20 cm radius which can be placed inside a 55-gallon drum (see Figure 2). Thus, the standards were constructed similarly to the ^{239}Pu standards in that any self-attenuation[10] or end effects issues are either eliminated or reduced to the point of being negligible.

Three ^{235}U (enriched to 92.11%) drum standards, consisting of 33.199, 100.987, and 200.428 grams respectively of ^{235}U, were designed and fabricated in 1992.[7] The design was aided by a Monte Carlo study with particular emphasis on eliminating self-absorption due to particle size and end effects. Each drum consists of twenty 4-liter polyethylene bottles containing the uranium disbursed within a diatomaceous earth matrix. The bottles were arranged within each drum in three layers of six bottles per layer (cylindrical symmetry with 60 degree radial separation) with the final two in the center. The bottles were held in position by a Cellutex matrix. Cellutex was chosen because the density (0.26 g/cm3) is almost identical to that of diatomaceous earth and has similar transmission properties. Thus, here as with the two sets of plutonium standards described above, the concerns expressed in reference 10 concerning calibrating using Pu standards should be greatly mitigated in this particular study.

We next focus our attention on the filtering and collimation geometry. In this type of work, the plutonium spectrum typically has a very intense ^{241}Am gamma-ray peak at 59.54 keV. Left unattended, that peak can take up a sizable portion of the count rate of the entire spectrum thereby considerably degrading the counting efficiency because of the increased dead time. Because of its low energy, placing a relatively low-Z filter in front of the detector collimator can easily reduce it. Care must be exercised because too much thickness filter can also reduce the low-energy plutonium peaks too much -- about 1/32 inch cadmium proved to be optimum for use with our standards.

Because we are comparing the TGS with an SGS set up with a 2-inch collimation geometry designed to measure 55-gallon drums, we configured the TGS accordingly. We used a 2-inch diamond-shaped collimator with trim pieces and set the collimator distance as close to the drum surface as possible – corresponds to a setting of about "70" on our shelf position indicator. We used a layer thickness of 2.3 inches, which corresponds to 15 layers for a normal-sized 55-gallon drum. In setting up the response matrices in TGS_MAT,[5] we used a layer coupling of 4 as derived from a simple geometric

calculation. We used a 10 X 10 grid, corresponding to 100 voxels per layer or about a 2-inch resolution. We were constrained by the ANTECH[6] MasterScan software (we used v 4.2.0 (Feb. 2003) throughout this study) to use 150 views per layer with the 100 voxels per layer setting. We used 600 ms per view, which seems to correlate best with the 2-inch resolution and which works out to about a 45-minute count time for a 15-layer 55-gallon drum. However, a 15-layer drum actually takes about 54 minutes because of the straight-through run time as well as time to position and change layers.

The MasterAnalysis software[6] (we used v 3.1.4 (May 2003)) allowed us to look at the transmission and emission spectra as well as the related images generated from the solutions to the matrix equations. In the transmission spectrum shown in Figure 3, the ^{109}Cd rate-loss peak is the large peak at the far left of the spectrum (88 keV). Because

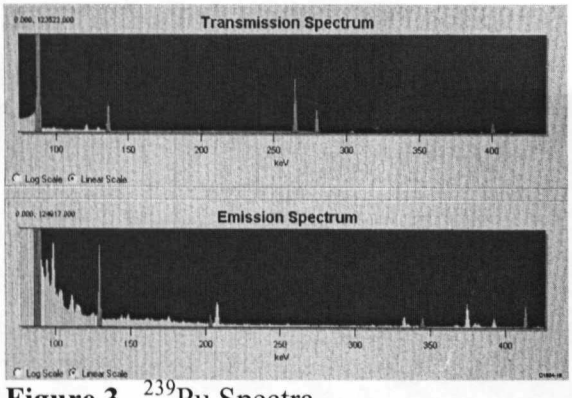

Figure 3. ^{239}Pu Spectra

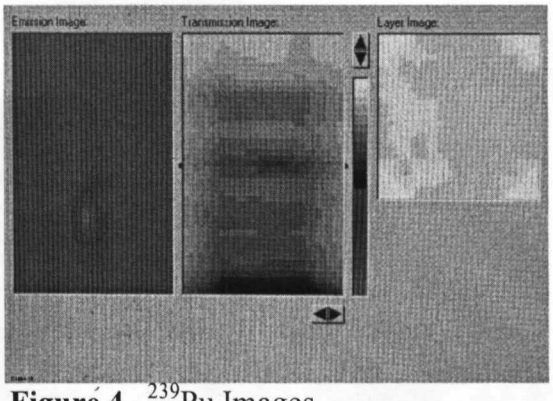

Figure 4. ^{239}Pu Images

the ^{109}Cd source is affixed to the detector, the peak also appears in the emission spectrum. The remaining four peaks at higher energy are from the ^{75}Se transmission source. The primary transmission peak used in this work is that at 400.66 keV seen at the far right of the spectrum. Four ^{239}Pu peaks are seen in the emission spectrum with the primary at 413.7 keV at the far right of the spectrum. In Figure 4 we see the emission and transmission images. Because the ^{239}Pu sheet standard was reasonably well localized and placed in an approximate 9 inch by 11 inch overpack container, it appears as a bright image roughly centered in the drum. We can see the platform as the dark area at the base of the transmission image. The somewhat dark area roughly centered in the transmission image probably corresponds to the lid of the overpack container. The layer image at the far right in Figure 4 is a vertical view at, in this case, roughly the center of the drum.

Figures 5 and 6 show the ^{238}Pu transmission and emission spectra and images. The transmission spectra are the same in all cases. We used the 153 keV emission peak, which is third from the left in Figure 5. It is interesting to note that two peaks at very much higher energy, especially the one at 766 keV, might be useful for dense matrices. However, the transmission source would probably have to be changed to one having higher energy peaks.[11] We used only two of the twelve rods and it appears that most of the ^{238}Pu was located at the top and bottom of the rods. The base of the carousel can be seen as the slightly darker image above the dark platform in the transmission image.

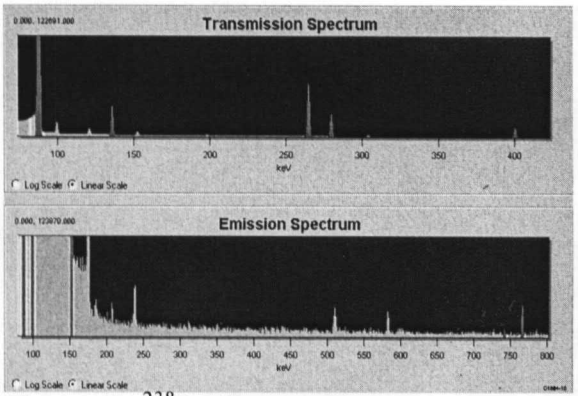

Figure 5. ^{238}Pu Spectra

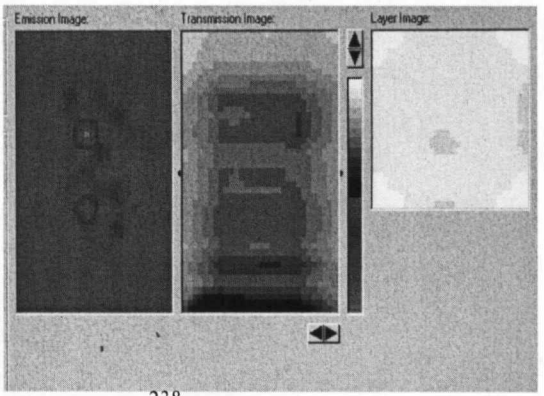

Figure 6. ^{238}Pu Images

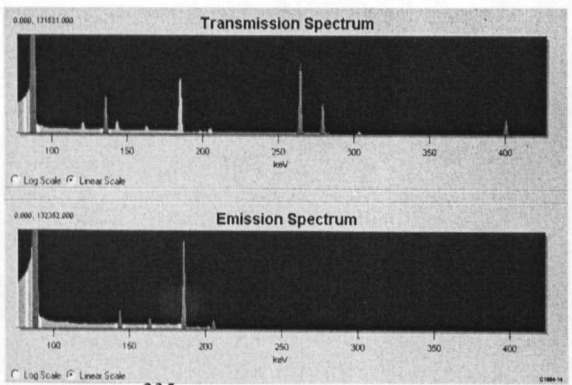

Figure 7. ^{235}U Spectra

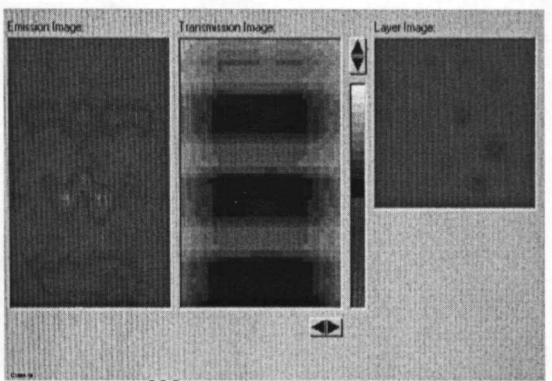

Figure 8. ^{235}U Images

Figures 7 and 8 show the ^{235}U transmission and emission spectra and images. We used the 186 keV emission peak. Here, unfortunately, the highest energy peak in the entire spectrum is the relatively weak one at 205 keV so this imposes a limitation on the usefulness of this technique for very dense ^{235}U matrices. The images are very interesting. We can clearly see that the poly bottles containing the uranium oxide diatomaceous earth mixture are arranged in layers with cylindrical symmetry, as described earlier. The layer image this time is an emission image where again the cylindrical symmetry is apparent.

Results and Discussion

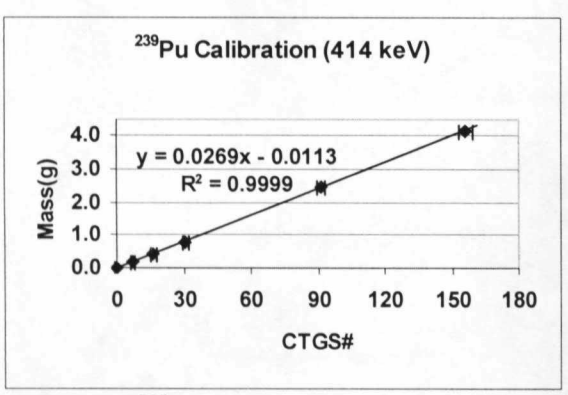

Chart 1. ^{239}Pu Calibration (TGS)

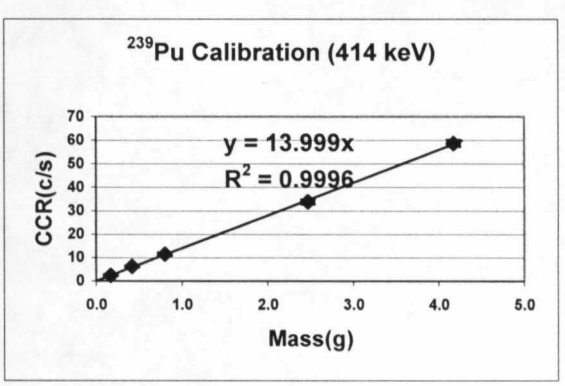

Chart 2. ^{239}Pu Calibration (SGS)

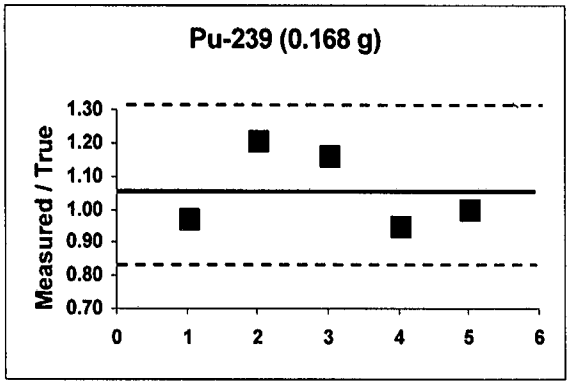

Chart 3. ^{239}Pu TGS Results

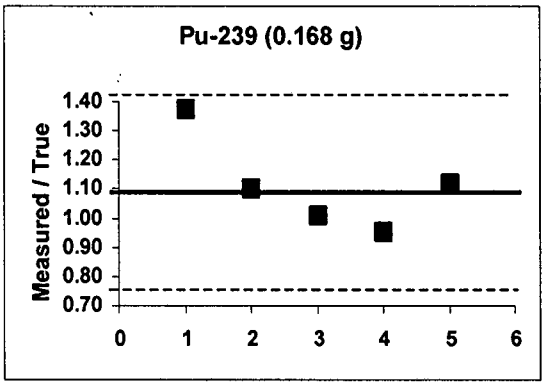

Chart 4. ^{239}Pu SGS Results

The calibration results shown in the first two charts seem to be quite good. The mass is plotted as a function of TGS# corrected for dead time and attenuation for the TGS – CCR for the SGS is count rate similarly corrected. The error bars are barely visible and the correlation coefficients for the linear fits are good. We show five replicate assays on our lowest mass standard in Charts 3 and 4. The dashed lines show the approximate two-sigma levels. There is a small positive bias on both instruments for these five replicate assays, which isn't too surprising considering the very small mass being measured. As expected, the TGS performed slightly better, as it did on the other ^{239}Pu standards as well.

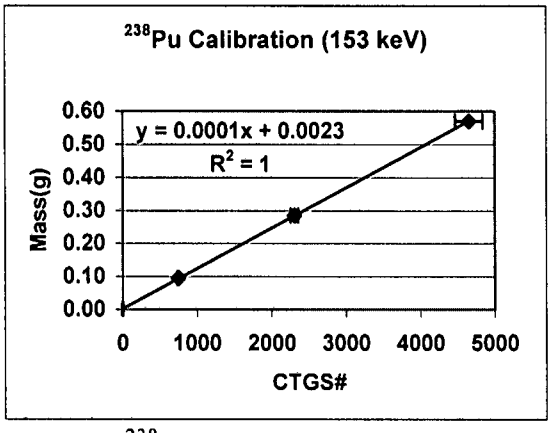

Chart 5. ^{238}Pu Calibration (TGS)

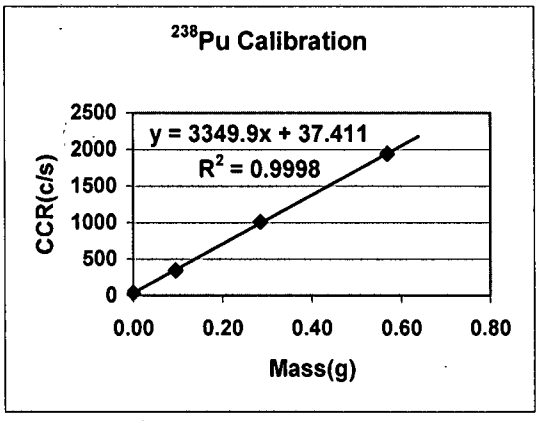

Chart 6. ^{238}Pu Calibration (SGS)

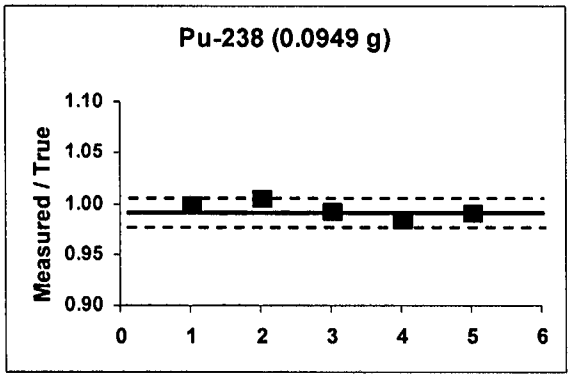

Chart 7. ^{238}Pu TGS Results

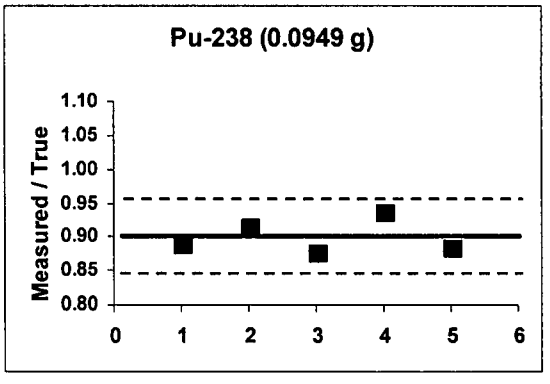

Chart 8. ^{238}Pu SGS Results

Again, for ^{238}Pu the calibration results appear good at first glance. The $R^2 = 1$ for the TGS us undoubtedly due to round-off. However, there is a rather large offset for the SGS calibration, which might have helped cause the rather large bias seen in the five replicate run results. The TGS results not only exhibit no bias but also show a much tighter grouping. Results for the other two ^{238}Pu standards were similar.

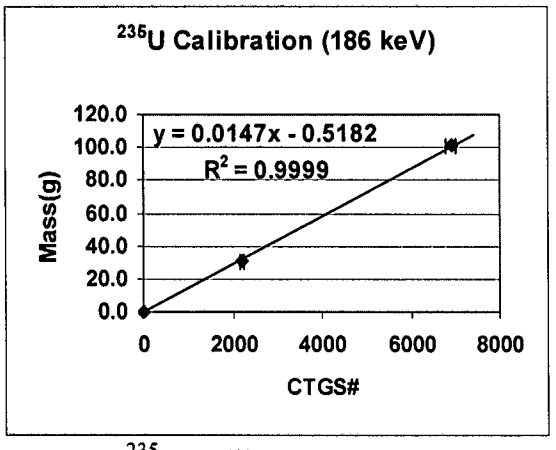

Chart 9. ^{235}U Calibration (TGS)

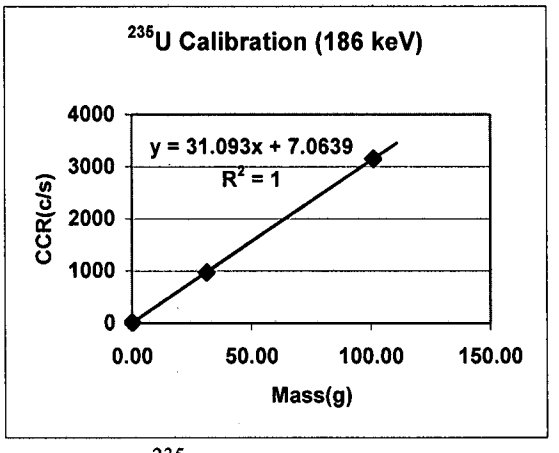

Chart 10. ^{235}U Calibration (SGS)

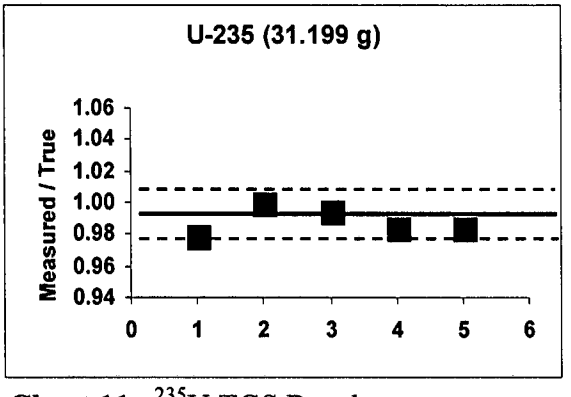

Chart 11. ^{235}U TGS Results

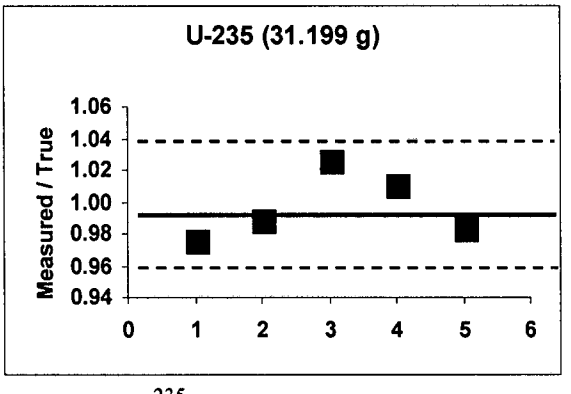

Chart 12. ^{235}U SGS Results

Here again, the calibrations look good except that they were done on only two uranium drum standards – the only two we had available at the time. Care must be exercised to ensure that sufficient data are taken on each standard to minimize error in the slope of the calibration curve. There is almost no bias on either instrument but the precision certainly looks better on the TGS for these five replicate assays.

Conclusions

In an effort to understand the performance characteristics of our newly acquired TGS, calibration and assay data were acquired on three different sets of standards differing widely in matrix and geometry. These results were compared with similar data from the same standards on our SGS instrument. To our knowledge, this is the first time such a comparison study on identical standards has been undertaken. The results were not

radically different, as expected, because the standards were ideally fabricated for the SGS. Nevertheless, while the accuracies were approximately equivalent (except for the ^{238}Pu case where the SGS calibration was probably somewhat off), the precision on five replicate assays on the smallest mass standards was better on the TGS in all cases. However, the real test will come in the near future when we measure items in complicated, heterogeneous matrices where the superior attenuation-correcting capability of the TGS will be of paramount importance.

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